

Repulsion-to-attraction transition in correlated electron systems triggered by a monocycle pulse

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We study the time evolution of the Hubbard model driven by a half-cycle or monocycle pulsed electric field $F(t)$ using the nonequilibrium dynamical mean-field theory. We find that for properly chosen pulse shapes the electron-electron interaction can be effectively and permanently switched from repulsive to attractive if there is no energy dissipation. The physics behind the interaction conversion is a nonadiabatic shift δ of the population in momentum space. When $\delta \sim \pi$, the shifted population relaxes to a negative-temperature state, which leads to the interaction switching. Due to electron correlation effects δ deviates from the dynamical phase $\phi = \int dt F(t)$, which enables the seemingly counterintuitive repulsion-to-attraction transition by a monocycle pulse with $\phi = 0$.

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I. INTRODUCTION

Controlling the interparticle interactions in a correlated electron system by applying intense laser fields is a challenging and exciting perspective, which may lead to states of matter that do not exist in equilibrium. For example, if one could effectively change the electron-electron interaction from the original Coulomb repulsion to an attraction, this may induce an s -wave superconducting state with very high transition temperature (optimally ≈ 0.1 bandwidth),^{1,2} or the BCS-BEC crossover.^{2,3} It will also enable us to study phenomena characteristic of nonequilibrium quantum systems, such as transient states after an interaction quench.⁴⁻⁶ The control of interparticle interactions is in fact possible in cold-atom systems,⁷ where one can manipulate the interaction in a wide range from repulsive to attractive using the Feshbach resonance that dominates the scattering length,⁸ but such a technique cannot be applied to electron systems.

One way to control the interaction is to create a population inversion in metallic bands corresponding to a negative-temperature (T) state.^{9,10} This implies an effective switching of the interaction from repulsive to attractive, since a density matrix $e^{-H/T}$ for a Hamiltonian H with temperature $T < 0$ corresponds to the one for the inverted $-H$ with $-T > 0$.^{11,12} While a (partial) population inversion itself is a common phenomenon (e.g., in laser productions), the interaction conversion is a genuine correlation effect in nonequilibrium. Ideally, the laser fields that drive the system should be “pulsed” waves since, first, the available intensity is generally much higher for ultrafast pulses¹³⁻¹⁵ than for continuous-wave lasers, and second, continued heating can be avoided. These considerations raise a fundamental question: can irradiation by a single-cycle pulse put a system into a negative- T state that survives for a long time after the pulse?

In this paper, we show that it is possible to induce a population inversion in metallic systems using a properly shaped monocycle or half-cycle pulse, and that in the absence of energy dissipation, the system will thermalize in the negative- T state after the pulse. By solving the driven Hubbard model with the nonequilibrium dynamical mean-field theory (DMFT),^{16,17} we will demonstrate that pulse fields $F(t)$ with

proper asymmetry between the positive [$F(t) > 0$] and negative [$F(t) < 0$] parts trigger a repulsion-to-attraction transition. Such asymmetric pulses can readily be generated thanks to the recent progress in laser techniques,¹⁸⁻²⁰ while their potential application to correlated systems has remained unexplored, in contrast to symmetric, many-cycle pulses.^{21,22}

Our strategy is to induce a nonadiabatic shift (denoted by δ) in the momentum distribution of the electrons by the asymmetric monocycle pulse (see Fig. 1). If we can achieve $\delta \simeq \pi$ (half of the Brillouin zone), the system is brought to a negative- T state [Fig. 1(c)], which amounts to a change of the interaction from repulsive to attractive. In a one-body picture, one expects that each electron acquires from the pulse field a dynamical phase

$$\phi = \frac{ea}{\hbar} \int_{-\infty}^{\infty} dt F(t) \quad (1)$$

with e the elementary charge and a the lattice constant (hereafter we set $e = a = \hbar = 1$). This causes a momentum shift $k \rightarrow k + \phi$, so that we simply have $\delta = \phi$. An immediate question is: can a monocycle pulse with $\int dt F(t) = 0$ (as dictated by Maxwell’s equation²³) induce a nontrivial shift of the population? We show that it is in fact possible in interacting systems, where the nonadiabatic shift δ exhibits a clear deviation from ϕ due to correlation effects. This allows us to achieve $\delta \simeq \pi$ even when $\int dt F(t) \propto \phi = 0$ by choosing the pulse shape appropriately. We reveal conditions for the pulse shape that lead to the population inversion, and construct a ‘phase diagram’ for the pulse-driven Hubbard model. We emphasize that the interaction conversion robustly persists after

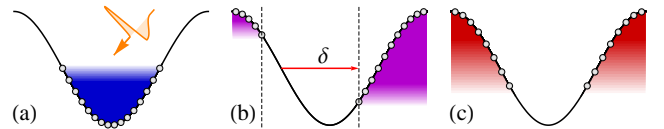


FIG. 1. (Color online) Schematic band pictures for the pulse-induced phase shift. (a) Initial system in equilibrium, (b) right after the pulse excitation, with the population shifted in momentum space by δ , and (c) the system finally thermalized with a negative temperature when $\delta \simeq \pi$.

the pulse has passed, at least in an isolated system without energy dissipation. This contrasts with the previously proposed scenario for the repulsion-to-attraction transition using continuous-wave fields.¹²

II. MODEL AND METHOD

We take, as the simplest model for correlated electrons, the single-band Hubbard model driven by an electric field with the Hamiltonian

$$H(t) = \sum_{ij,\sigma} t_{ij} \exp \left(-i \int_{\mathbf{R}_j}^{\mathbf{R}_i} d\mathbf{r} \cdot \mathbf{A}(t) \right) c_{i\sigma}^\dagger c_{j\sigma} + H_{\text{int}}(U), \quad (2)$$

where t_{ij} is the hopping between sites at \mathbf{R}_i and \mathbf{R}_j , the electric field $\mathbf{F}(t) = -\partial \mathbf{A}(t)/\partial t$ is expressed in terms of the vector potential $\mathbf{A}(t)$, and c^\dagger (c) creates (annihilates) an electron. For the interaction we take the particle-hole symmetric form

$$H_{\text{int}}(U) = U \sum_i \left(n_{i\uparrow} - \frac{1}{2} \right) \left(n_{i\downarrow} - \frac{1}{2} \right), \quad (3)$$

where $U (\geq 0)$ is the repulsive Coulomb interaction with $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$. We apply a pulsed wave at $t = 0$, and switch off the field at $t = \tau$. For the DMFT, we consider a hypercubic lattice with the Gaussian density of states $D(\epsilon) = \frac{1}{\sqrt{\pi}W} e^{-\epsilon^2/W^2}$,¹⁶ and apply the field in the diagonal direction with $\mathbf{F}(t) = F(t)(1, 1, \dots)$. The band is assumed to be half-filled. Throughout the paper, we use the bandwidth W as the unit of energy, and take the initial temperature to be $T = 0.1$.

III. RESULTS

A. Noninteracting system

Let us start with the noninteracting system. We focus on the momentum distribution defined by $f(\mathbf{k}, t) = -i\tilde{G}_{\mathbf{k}}^<(t, t) = -i\tilde{G}_{\mathbf{k}+\mathbf{A}(t)}^<(t, t)$, where $G_{\mathbf{k}}^<(t, t')$ [$\tilde{G}_{\mathbf{k}}^<(t, t')$] is the (gauge-invariant²⁴) lesser Green function. For the noninteracting system, the lesser Green function is given by

$$G_{0\mathbf{k}}^<(t, t') = i f_0(\epsilon_{\mathbf{k}}) \exp \left(-i \int_{t'}^t d\tilde{t} \epsilon_{\mathbf{k}-\mathbf{A}(\tilde{t})} \right), \quad (4)$$

where $f_0(\epsilon) = 1/(e^{\epsilon/T} + 1)$ is the Fermi distribution, and $\epsilon_{\mathbf{k}}$ the band dispersion. After the pulse excitation ($t > \tau$), the momentum distribution becomes $f(\mathbf{k}, t) = f_0(\epsilon_{\mathbf{k}-\phi})$ with $\phi = -\mathbf{A}(\tau) = \phi(1, 1, \dots)$. Note that the effect of the pulse field on the final state amounts to a momentum shift ϕ (1). For a π shift ($\phi \simeq \pi$), the electrons occupy the band top with $f(\mathbf{k}, t) \sim f_0(-\epsilon_{\mathbf{k}})$, which is characterized by an effective temperature $T_{\text{eff}} = -T < 0$. Thus a π shift (which may be viewed as a partial Bloch oscillation) is the condition that leads to a negative- T state in the noninteracting system.

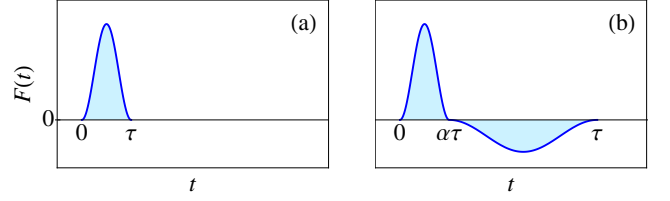


FIG. 2. (Color online) Schematic temporal profiles of a half-cycle pulse (a) and a monocycle pulse (b) for which α controls the asymmetry.

B. Interacting system

Now let us move on to the interacting case. There, we can identify the repulsion-to-attraction transition from the total energy $E_{\text{tot}}(t) = \langle H(t) \rangle$: After the pulse excitation, a (nonintegrable) isolated system is supposed to approach a thermalized state²⁵ with some effective temperature T_{eff} and a total energy $E_{\text{tot}}(\tau)$ (which is conserved after the pulse is over at $t = \tau$). A thermal state with a positive temperature always gives $E_{\text{tot}} < 0$ at half filling for the interaction term (3), while one with a negative temperature gives $E_{\text{tot}} > 0$. This suggests that the total energy plays the role of an “order parameter” for the repulsion-to-attraction transition. If and only if $E_{\text{tot}}(\tau) > 0$ the system arrives at a negative- T state ($T_{\text{eff}} < 0$), in which case the density matrix is given by

$$\begin{aligned} \rho &\propto \exp \left(-\frac{1}{T_{\text{eff}}} \left[\sum_{\mathbf{k},\sigma} \epsilon_{\mathbf{k}+\phi} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + H_{\text{int}}(U) \right] \right) \\ &= \exp \left(-\frac{1}{|T_{\text{eff}}|} \left[\sum_{\mathbf{k},\sigma} \epsilon_{\mathbf{k}} \tilde{c}_{\mathbf{k}\sigma}^\dagger \tilde{c}_{\mathbf{k}\sigma} + \tilde{H}_{\text{int}}(-U) \right] \right). \end{aligned} \quad (5)$$

Here we have introduced a gauge transformation

$$c_{i\sigma} \rightarrow \tilde{c}_{i\sigma} = e^{-i(\phi+\pi)(1,1,\dots) \cdot \mathbf{R}_i} c_{i\sigma}$$

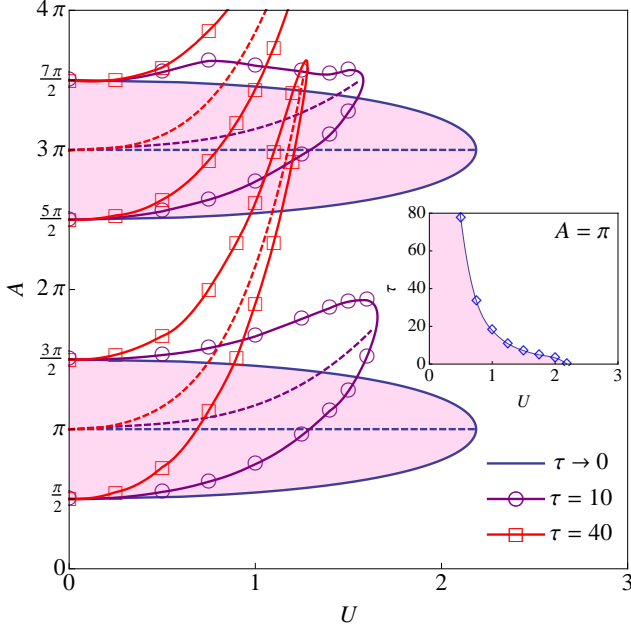
with $\tilde{H}_{\text{int}}(-U) = -U \sum_i (\tilde{n}_{i\uparrow} - \frac{1}{2})(\tilde{n}_{i\downarrow} - \frac{1}{2})$ (with $\tilde{n}_{i\sigma} = \tilde{c}_{i\sigma}^\dagger \tilde{c}_{i\sigma}$) to cancel the phase shift in the kinetic energy. The above equation implies that the state can be viewed as a thermal state with a positive T and an attractive interaction $-U < 0$. This is the basic mechanism behind the repulsion-to-attraction transition driven by the pulse. Note that the condition for the interacting system is $T_{\text{eff}} < 0$, as opposed to the noninteracting counterpart $\phi \simeq \pi$.

To make our argument more precise, we consider two types of pulses. One is a half-cycle pulse [Fig. 2(a)], and the other is a monocycle pulse [Fig. 2(b)]:

$$F_{\text{half-cycle}}(t) = \frac{A}{\tau} s\left(\frac{t}{\tau}\right), \quad (6)$$

$$F_{\text{monocycle}}(t) = \frac{A}{\alpha\tau} s\left(\frac{t}{\alpha\tau}\right) - \frac{A}{(1-\alpha)\tau} s\left(\frac{\tau-t}{(1-\alpha)\tau}\right). \quad (7)$$

Here A controls the amplitude of the pulse, $s(x) (\geq 0)$ is a pulse shape function that has support in $0 \leq x \leq 1$ with $\int_0^1 dx s(x) =$



1, and α ($0 < \alpha < 1$) controls the asymmetry of the monocycle pulse, with $\alpha = \frac{1}{2}$ corresponding to the symmetric case [$F(\tau - t) = -F(t)$]. The dynamical phase (1) is $\phi = A$ ($\phi = 0$) for the half- (mono-)cycle pulse.

C. Half-cycle pulse

We first consider the half-cycle pulse (6). The simplest case is the limit $\tau \rightarrow 0$, corresponding to a delta-function pulse [$F(t) \rightarrow A\delta(t)$]. In this case, the momentum shift is $\delta = \phi$, so that the order parameter reads

$$E_{\text{tot}}(\tau) = E_{\text{kin}}(0) \cos \phi - iJ(0) \sin \phi + E_{\text{int}}(0),$$

where $E_{\text{kin}}(t)$, $J(t)$, $E_{\text{int}}(t)$ are the kinetic energy, current, and interaction energy at time t , respectively. Since $J(0) = 0$ in the initial state and $\phi = A$ for the half-cycle pulse (6), we can identify the condition for the repulsion-to-attraction transition,

$$E_{\text{kin}}(0) \cos A + E_{\text{int}}(0) > 0, \quad (8)$$

which is completely determined by the equilibrium state. The criterion is quite general, so should be applicable to systems with any fillings in any dimensions if one puts the origin of the energy to be the one at $T = \pm\infty$. In Fig. 3, we show the attractively interacting regions by the hashed areas. Attractive regions appear periodically in the amplitude A of the pulse as a series of lobes around $A \simeq (2n + 1)\pi$ ($n = 0, 1, 2, \dots$). Each

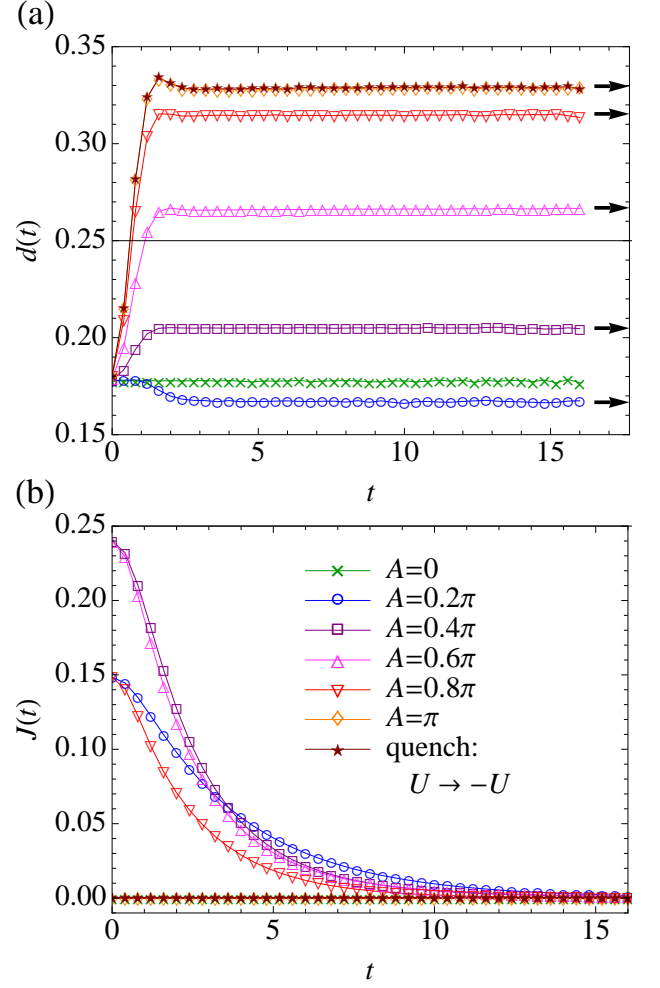


FIG. 4. (Color online) Time evolution of the double occupancy (a) and the current (b) after the delta-function-pulse excitation, which are compared with the interaction quench ($U \rightarrow -U$) with $U = 1$. Each arrow indicates the double occupancy in the corresponding thermal state with the same total energy.

lobe has the tip at $U_c = 2.186$, which turns out to be smaller than the critical U for the Mott transition,¹⁶ so that the transition always occurs in the metallic regime. The repulsion-to-attraction conversion is obviously distinct from a heating effect, since it appears and disappears repeatedly as one increases the amplitude A of the pulse field.

To study how the system evolves in time, we have numerically solved the model (2) with the nonequilibrium DMFT.¹⁷ As an impurity solver for DMFT, we mainly employ the continuous-time quantum Monte Carlo method²⁶ with the weak-coupling expansion generalized to nonequilibrium.²⁷ To capture the long-time ($t \geq 20$) behavior in the very weak-coupling regime ($U \leq 1.2$) we use the iterative perturbation theory,^{16,28} which is known to give quite accurate results up to a long time²⁹ for such small U at half filling.

In Fig. 4(a) we show how the double occupancy $d(t) \equiv \langle n_{\uparrow} n_{\downarrow}(t) \rangle$, a measure of the interaction, evolves after the delta-

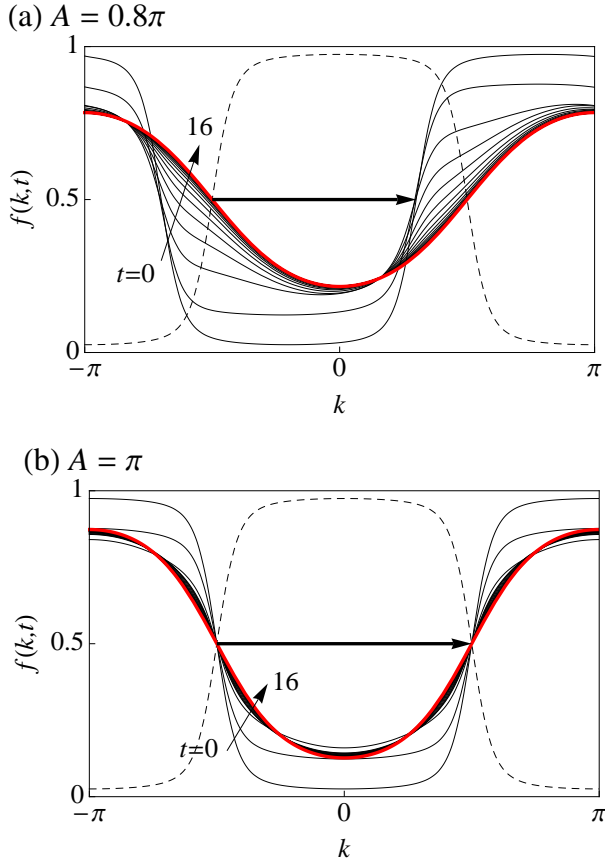


FIG. 5. (Color online) Snapshots of the corresponding momentum distribution $f(k, t)$ ³⁰ (black curves) at $t = 0, 0.8, 1.6, \dots, 16$ are shown for $A = 0.8\pi$ (a) and $A = \pi$ (b). They converge to thermal distributions with $T_{\text{eff}} = -0.725$ and $T_{\text{eff}} = -0.446$ (thick red curves), respectively. Dashed curves represent the initial distributions. The horizontal arrows indicate the pulse-induced phase shift $\delta = \phi = A$.

function pulse in an initially repulsive system ($U = 1$). We notice that for $A > 0.5\pi$ $d(t)$ shoots well beyond the noninteracting value $d = \langle n_{\uparrow} \rangle \langle n_{\downarrow} \rangle = 0.25$, which implies that the electrons do indeed start to attract each other after the pulse, as predicted from the criterion (8). The repulsion-to-attraction transition is “perfect” for $A = \pi$, where the temporal evolution of d is found to accurately agree with that for the interaction quench, $U \rightarrow -U$ [Fig. 4(a)]. For this “ π pulse”, the shift of the momentum just changes the sign of the hopping ($\epsilon_k \rightarrow \epsilon_{k+\phi} = -\epsilon_k$), which is known to be equivalent to interaction quench.¹²

Remarkably, after the pulse excitation $d(t)$ relaxes quickly to a steady state ($t \lesssim 3$). We have confirmed that it converges to the thermal value d_{th} [indicated by arrows in Fig. 4(a)] for the equilibrium state having the same E_{tot} . For $A > 0.5\pi$ the corresponding temperature (T_{eff}) of the thermal state is negative since $E_{\text{tot}} > 0$. Note that d_{th} is a nonmonotonic function of temperature, so that $d(t)$ decreases in time for $A = 0.2\pi$.

On the other hand, the current $J(t)$ [Fig. 4(b)] generated by the momentum shift $k \rightarrow k + \phi$ decays more slowly ($t \lesssim 15$) for $0 < A < \pi$ than $d(t)$. The slow relaxation is also seen in

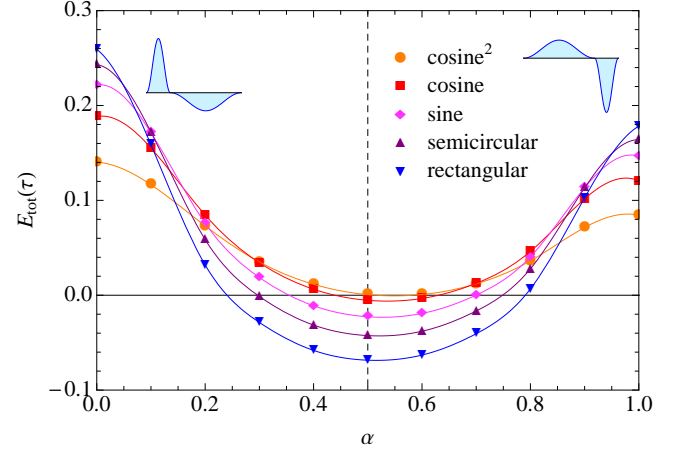


FIG. 6. (Color online) The total energies at $t = \tau$ for the system with $U = 1$ driven by monocycle pulses with $A = \pi$, $\tau = 40$ plotted for various pulse shapes as a function of the asymmetry parameter α .

the momentum distributions $f(k, t)$ ³⁰ [Fig. 5(a) for $A = 0.8\pi$ and (b) for $A = \pi$]. A similar separation of the relaxation times of $d(t)$ and $f(k, t)$ has been observed in the interaction quench,⁶ and was attributed to the existence of a “prethermalized” state.⁵ Here the slow decay becomes particularly evident when the momentum shift is not perfect (i.e., $A \neq \pi$). In this case the system needs to adjust the momentum shift to π to achieve a thermal state. Since the relaxation involves a violation of the momentum conservation by Umklapp scattering, it takes longer than the relaxation of the double occupancy via particle-hole annihilations. The distributions eventually relax to thermal states with $T_{\text{eff}} < 0$ [red curves in Fig. 5(a), (b)].

So far we have examined the delta-function pulse ($\tau \rightarrow 0$). To be more realistic it is important to evaluate the effect of τ on the transition. Here we take, as an example, a half-cycle pulse with

$$s(x) = 1 - \cos(2\pi x), \quad (9)$$

which we call the “cosine pulse”. We can again use $E_{\text{tot}}(\tau)$ as an indicator for the interaction conversion. In the inset of Fig. 3, we show how the critical interaction (U_c) of the repulsion-to-attraction transition induced by the cosine pulse with $A = \pi$ depends on τ . For $\tau \lesssim 10$, $U_c(\tau)$ rapidly falls off from $U_c(\tau = 0)$, while for larger τ it decays to zero very slowly. In the adiabatic limit ($\tau \rightarrow \infty$) $E_{\text{tot}}(\tau) \rightarrow E_{\text{tot}}(0) < 0$ for $U > 0$ and the repulsion-to-attraction transition naturally disappears.

For general A and non-zero τ of the cosine pulse, Fig. 3 depicts the phase diagram for the pulse-driven Hubbard model. The attractive regions now deform in a characteristic manner, i.e., the tips of the lobes bend toward larger A , which becomes drastic for $\tau = 40$. The deformation of the phase diagram implies a rather counterintuitive fact: for $\tau = 40$ and $U \sim 0.9$ the repulsion-to-attraction conversion occurs even for a trivial phase $\phi = A = 2\pi$. This suggests that the effective phase shift δ that the correlated system acquires is not equal to ϕ for nonzero τ . Using the extremal points of the total energy

satisfying $\partial E_{\text{tot}}(\tau)/\partial A = 0$ (dashed curves in Fig. 3) as an estimate for $\delta = (2n + 1)\pi$, we can see the large deviations of δ from $\phi = A = (2n + 1)\pi$ as U and τ grow. We attribute this to a correlation effect: During irradiation with the pulse the electrons scatter with each other, which causes broadening of the momentum distribution. Consequently the shift in the momentum is suppressed, and δ becomes smaller than ϕ .

D. Monocycle pulse

The result that $\delta \neq \phi$ suggests an experimentally much simpler way to induce the repulsion-to-attraction transition ($\delta \simeq \pi$) by monocycle pulses (7) with $\phi = 0$. The basic idea is the following: since the effect of a half-cycle pulse very much depends on its width, we can suitably choose the widths of the first and second half cycles of a monocycle pulse so that the total phase shift is $\simeq \pi$. In Fig. 6, we plot the total energy at $t = \tau$ for various types of pulse shapes. The shape function of each pulse is defined by

$$s(x) = \begin{cases} \frac{2}{3}[1 - \cos(2\pi x)]^2 & \text{cosine}^2 \text{ pulse,} \\ \frac{\pi}{2} \sin(\pi x) & \text{sine pulse,} \\ \frac{4}{\pi} \sqrt{1 - (2x - 1)^2} & \text{semicircular pulse,} \\ 1 & \text{rectangular pulse,} \end{cases}$$

for $0 \leq x \leq 1$. $s(x)$ of the cosine pulse is defined as before [Eq. (9)]. From the above argument, the asymmetry, here represented by α , should be important, and we can indeed see that $E_{\text{tot}}(\tau)$ becomes positive (implying a repulsion-to-attraction transition) as soon as we go sufficiently away from the symmetric pulse form ($\alpha = \frac{1}{2}$). To be more precise, the momentum shift induced by the first half cycle does not cancel the one induced by the second half cycle when the mono-pulse is sufficiently asymmetric. Note that the cosine² pulse gives $E_{\text{tot}}(\tau) > 0$ even at $\alpha = \frac{1}{2}$, although its value is very small. There is a slight difference in $E_{\text{tot}}(\tau)$ for the cases with $\alpha < \frac{1}{2}$ (i.e., the sharp pulse comes first, followed by the broad one) and $\alpha > \frac{1}{2}$, implying that the former is more suitable than the latter to induce the attractive interaction. The order parameter $E_{\text{tot}}(\tau)$ also depends on the shape of the pulse. In the

case of $U = 1$ and $\tau = 40$, the larger the peak amplitude [$s(\frac{1}{2}) = \frac{8}{3}, 2, \frac{\pi}{2}, \frac{4}{\pi}, 1$ for the cosine², cosine, sine, semicircular, rectangular pulse, respectively], the larger the $E_{\text{tot}}(\tau)$ around $\alpha = \frac{1}{2}$. For even shaper pulse shapes (cosine³, ...), $E_{\text{tot}}(\tau)$ at $\alpha = \frac{1}{2}$ starts to decrease, so that the cosine² pulse is an optimal shape in this case.

IV. DISCUSSION AND OUTLOOK

Finally, let us discuss the experimental feasibility of the pulse-induced repulsion-to-attraction transition proposed here. Asymmetric monocycle pulses with qualitative features comparable to the shapes considered here can be generated experimentally.³¹ One way to detect the negative- T state is to measure the time-resolved dc or optical conductivity, which will become negative after the pulse irradiation due to energy gain. Another possibility is to measure momentum-resolved photoemission spectra, which can detect the shift in the momentum distribution [Fig. 5(a), (b)]. We require the time resolution of the measurement to be fine enough that it can detect the population-inverted state before it relaxes to a more stable state through energy dissipation. The dissipation typically occurs due to phonons whose time scale is of the order of 0.1–1 ps,³² which allows one to access the negative- T state using current ultrafast laser techniques with a resolution ~ 10 fs.¹⁴ Materials that have a metallic band at the Fermi energy, well separated from the other bands, are suitable candidates because of the absence of interband transitions that destabilize the population-inverted state. As an example, transparent conductors (e.g., Sn-doped In_2O_3 ³³) and alkali-metal-loaded zeolites³⁴ such as sodalite³⁵ are materials that exhibit this kind of band structure.

V. ACKNOWLEDGMENTS

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